Thermodynamics of a dense lead plasma near the high-temperature boiling curve

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Thermal measurements have been used for the first time to study the thermodynamics of a dense metal plasma in the critical region. Measurements of the temperature on rarefaction isentropes of lead samples of normal density shock-compressed to states with pressures of 1 and 1.6 Mbar are reported. The pressures at which the isentropes go into the two-phase liquid-vapor region have been measured.

The behavior of intensely heated liquid metals in the region of the phase diagram which lies between the solid state and the neutral vapor has recently attracted increased interest[1] because of both the fundamental interest in this topic and the many applications associated with the pulsed generation of a high, local concentration of energy. The occurrence of an intense and structurally complex interparticle interaction in this region, under conditions of disorder in the system, seriously hinders the use of reliable theoretical methods. As a result, there has been fertile ground for the development of a variety of hypotheses with regard to the qualitative form of the phase diagram of metals with parameter values well in the nonideal region.

For metals, the expected critical temperatures[2] are comparable to the ionization potentials, so the high-temperature evaporation of metals leads directly to a nonideal

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plasma phase; the region of a neutral vapor is skipped over, in contrast with the behavior of classical liquids. Zel'dovich and Landau[3] were apparently the first to point out that this behavior could result in some additional phase transitions which would qualitatively distort the conventional path of the boiling curve in its high-temperature region. Several hypotheses regarding the appearance of plasma[4] and cluster[5] phase transitions in the critical region of metals were also suggested in later studies. However, it is hardly possible to carry out an experimental study of dense, hot, plasma-metal states by conventional steady-state experimental methods, since the temperatures and pressures at the critical point for the overwhelming majority of metals (alkali metals and mercury are exceptions) go well beyond the heat resistance and strength of the structural materials used in experimental apparatus.[1]

In the present study we have used a dynamic method for generating these states in order to learn about the thermodynamics of an intensely heated liquid and a dense plasma of lead near the high-temperature boiling curve. This method is based on an isentropic expansion of the metal after it has been compressed and heated at the front of an intense shock wave.[6] In contrast with some previous papers[7,8] reporting studies by this method and containing results of only kinematic measurements, the present letter reports the results of the first thermal measurements in the critical region. The temperature measured in the rarefaction wave is much more sensitive than the expansion velocity to a metal evaporation process, since the transition from a single-phase state to the vapor-liquid region corresponds to a transition from a power-law to an exponential dependence of the equilibrium temperature on the pressure. Accordingly, the point of the slope change on the measured temperature dependence on the isentrope shows us the point at which it crosses the liquid-vapor equilibrium line. The lower part of the experimental curve determines the position of the evaporation line in the phase plane, since there is a single-valued relationship between the temperature and the pressure inside the two-phase region.

In order to produce intense shock waves in lead targets, we used high-velocity explosive linear shock-wave generators. The products of the detonation of condensed explosives accelerated strikers 60 mm in diameter made of steel (1 mm thick) or aluminum (2 mm) to velocities of 5.0 and 5.4 km/s, respectively. During the impact of these strikers on lead samples of normal density, shock waves with respective front pressures of 1.6 and 1 Mbar were excited. The velocity of the strikers and the intensity of the shock waves in the lead targets were monitored in a special series of experiments. The dimensions of the strikers and the targets were chosen in such a way that the effect of the rarefaction waves propagating from the back of the striker was eliminated.

The pressure in the isentropic expansion wave was detected by measuring the rarefaction velocity of shock-compressed lead in helium at various initial pressures. The low atomic weight and high ionization potential of helium make it transparent to the optical emission from the dense lead plasma. The plasma temperature was determined by measuring the optical emission with silicon photodiodes, positioned directly in the experimental apparatus, and with a two-channel pyrometer, working at wavelengths of 449 ± 5 nm and 560 ± 5 nm, which were singled out by interference filters. In these measurements we used SNFT-3 and ÉLU-31FK photomultipliers. To couple the light from the experimental apparatus to the photomultipliers, we used collimating optical fibers, which gave the measurement apparatus an excellent noise immunity and which allowed a precise localization of the surface region under study. The parameters were calibrated within an error of 3% with the help of a tungsten ribbon lamp at temperatures up to $T = 2800$ K. The electrical signals were measured by S9-4A high-speed oscilloscopes with a bandwidth of 0.5 GHz.

Figure 1 shows a typical oscilloscope trace of the intensity of the optical emission corresponding to moderate values of the expansion parameters of the metal into the region of the liquid phase. We see that the amplitude of the measured signal varies only slightly over the time interval 20–500 ns after the shock wave reaches the free surface. Consequently, the brightness temperature increases extremely insignificantly over the time scales of the dynamic experiment, $\sim 1\ \mu$s (an increase in the signal by 5–7% corresponds to a temperature increase of 1%). This behavior is evidence that the system under study is at thermodynamic equilibrium.[9]

The results of thermal measurements on the isentropes of the lead near the high-temperature boiling curve are shown in Fig. 2. Each experimental point here is the average of the results of two or three experiments, in each of which the brightness temperature was determined independently two or three times. The brightness temperatures which were measured were essentially identical. Figure 2 also shows data from static measurements for the saturated vapor[10] and results calculated for the temperature on the experimental isentropes and boiling curve on the basis of a wide-range equation of state for lead.[8] A comparison of the experimental data with the theoretical results according to the equation of state[8] constructed from the data of kinematic measurements[7,8] indicates that they are approximately the same and are similar at pressures $P \gtrsim 30$ bar. At lower pressures, on the other hand, we observe an increasing deviation of the experimental temperatures from the theoretical evaporation curve and the data of the static measurements. This deviation, which goes beyond the experimental error, apparently stems from a deviation of the expansion process from thermal dynamic equilibrium at low pressures and low densities. The lower limit on the applicability of the method of isentropic expansion established in this manner constitutes additional experimental confirmation of the results of an analysis of the

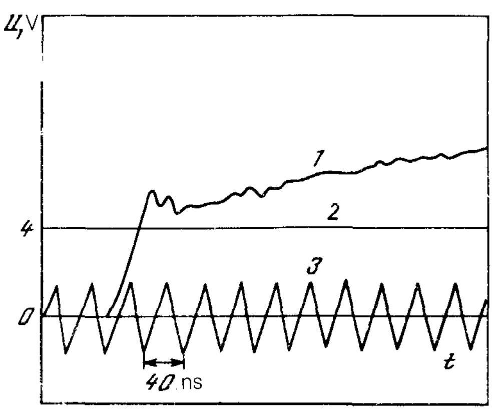

FIG. 1. 1—Typical oscilloscope trace of the optical emission intensity; 2—amplitude of the calibration signal, 4 V; 3—25-MHz sine-wave time reference.

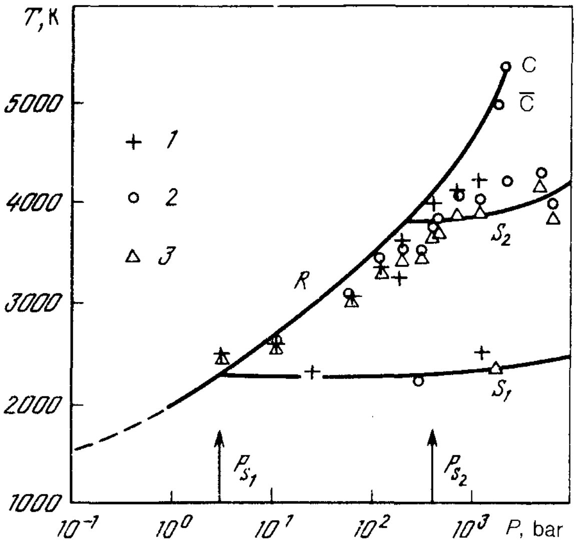

FIG. 2. Phase diagram of lead. Solid lines: Calculations[8] of the liquid-vapor equilibrium line (R) with the critical point (C) and the rarefaction isentrope (S). Dashed line: Static data on the vapor pressure of lead.[10] $\overline{\text{C}}$: estimate of the parameters of the critical point.[2] Points: Results of measurements of the temperature with silicon photodiodes (1) and with a two-channel pyrometer (2—$\lambda_1 = 449$ nm; 3—$\lambda_2 = 560$ nm). Here P_{s_1} bar and P_{s_2} are the pressures on the evaporation curve as the isentropes enter the two-phase region.

kinetics of the evaporation and condensation of metals in isentropic rarefaction waves.[9]

In order to learn about whether the thermodynamic states in the two-phase region are equilibrium states, we studied the expansion of lead from an initial state of shock compression at a pressure of 2.4 Mbar, achieved through the use of an explosive generator of elevated intensity. It was found experimentally that during the isentropic expansion of lead from various initial states, with $P = 1.6$ Mbar and $P = 2.4$ Mbar, to the same final state, with $P = 250$ bar, the measured temperatures are also equal at $T = 3700$ K. This result is evidence that the expansion of the lead in the two-phase region of the parameters is an equilibrium expansion, regardless of the value of the entropy. It constitutes further proof that the system is at thermodynamic equilibrium at the time scale of the process, $\sim 1\ \mu$s.

The thermal measurements which have been carried out, like the previous determination of kinematic parameters,[7,8] indicate that there are no significant thermodynamic anomalies which might be linked with phase transitions in a highly nonideal dense plasma[4,5] or with metal-insulator transitions in disordered metal structures.[3] The study which has been carried out makes it possible to detect the beginning of the evaporation of the metal fairly definitely, from the entry of the isentrope into the two-phase region. Another point, and a particularly important one, is that the curve of the high-temperature boiling of lead has been found in a single series of experiments over the broad pressure range $P = 50$–10^3 bar, up to pressures more than 10^3 times that in static measurements.[10]

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